

# Identifying Transition Metal Contribution to the Rare-Earth $L_2$ -Edge XMCD Spectra in R-T Intermetallics

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**Abstract.** We report on a transition metal contribution to the rare-earth  $L_2$ -edge XMCD spectrum of intermetallic rare-earth - transition metal compounds (R-T). The shape and location of this contribution is independent on both the specific rare-earth and the specific 3d metal in the compound. The evolution of the signal with the 3d content indicates that the signal is proportional to the number of T neighboring atoms around the absorbing R atom. In addition, comparison between Fe and Co Laves compounds indicates that the signal also resembles the magnetic moment of the T metal. Comparison with other R:T series shows that the presence of a T contribution is not a particularity of Laves phases but a general result.

**Keywords:** Intermetallic compounds, XAS, XMCD.

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## INTRODUCTION

The study of the rare-earth 5d states, by using the XMCD technique, is fundamental to the understanding of the magnetism of rare-earth (R) and transition metal (T) intermetallic compounds. However, the interpretation of the R  $L_{2,3}$ -edges XMCD spectra, which, in principle, should give us the magnetic characterization of R (5d) states, is not clear to date. This is due to the presence of contributions of quadrupolar origin and the spin dependence of the radial matrix elements of the dipolar transitions. [1-3] In a first approximation, the XMCD spectrum profile at these  $L_{2,3}$ -edges is considered as originating from the interplay between the R(5d) and R(4f) states. Moreover, recent theoretical works suggested the need of including the R(5d)-T(3d) hybridization to account for the R- $L_{2,3}$  XMCD in the case of R-T intermetallics [4]. Within this framework, we performed an XMCD study at the R  $L_{2,3}$ -edges through the  $R(Al_{1-x}Fe_x)_2$  series [5]. We found that as Al is replaced by Fe a new contribution emerges in the low energy region of the R  $L_2$ -edge spectrum. The evolution of this extra feature indicated that it was related to the Fe atoms.

On the other hand, if the simple presence of a magnetic 3d metal entails the emergence of a new contribution to the XMCD spectrum, a similar behavior should be expected independently on the intermetallic R-T compound. However, no experimental results have been reported in the case of Co compounds. In fact, previous works on  $ErT_2$  Laves phases do not indicate any difference in the profile of  $ErCo_2$  and  $ErAl_2$  compounds [6]. If our hypothesis is true, a similar profile and evolution should be found in the case of Laves phases of Co. Therefore, more experimental evidence is needed to verify if the presence of a contribution of 3d origin is a general result or a particularity of Fe Laves phases.

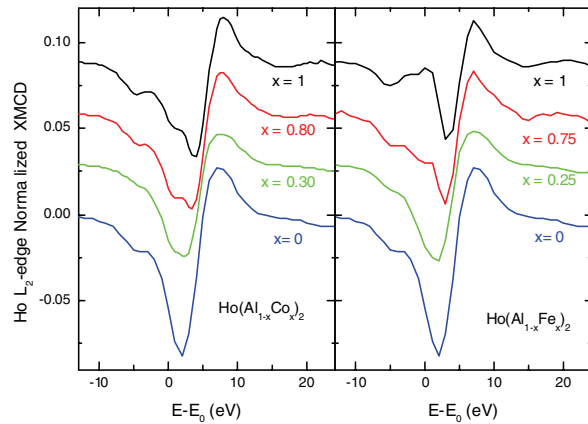
Trying to get a deeper insight into the magnetic information that can be obtained from these spectra, a systematic XMCD study at the Ho  $L_{2,3}$ -edges through the  $Ho(Al_{1-x}Co_x)_2$  series has been carried out. The comparison between the two, Fe and Co, series is expected to provide more complete information to interpret the features at the R  $L_2$ -edge. Moreover, as  $RCo_2$  and  $RFe_2$  present the same crystal structure, differences between these systems can only be related to electronic or magnetic effects.

## EXPERIMENTAL DETAILS

$\text{Ho}(\text{Al}_{1-x}\text{Fe}_x)_2$  and  $\text{Ho}(\text{Al}_{1-x}\text{Co}_x)_2$  samples were prepared by arc-melting the pure elements under Ar protective atmosphere. The ingots were annealed at 800-850°C for one week. The homogeneity of the compounds was checked by X-ray diffraction, which showed that all the samples were single phase. XMCD experiments were performed at the beamline BL39XU of the SPring-8 facility. XMCD spectra were recorded in the transmission mode using the helicity-modulation technique. [7] The XMCD spectra were recorded at  $T = 5$  K and under an applied magnetic field  $\mu_0 H = 5$  T. The spin-dependent absorption coefficient was then obtained as the difference of the absorption coefficient  $\mu_c = (\mu^- - \mu^+)$  for antiparallel,  $\mu^-$ , and parallel,  $\mu^+$ , orientation of the photon helicity and sample magnetization.

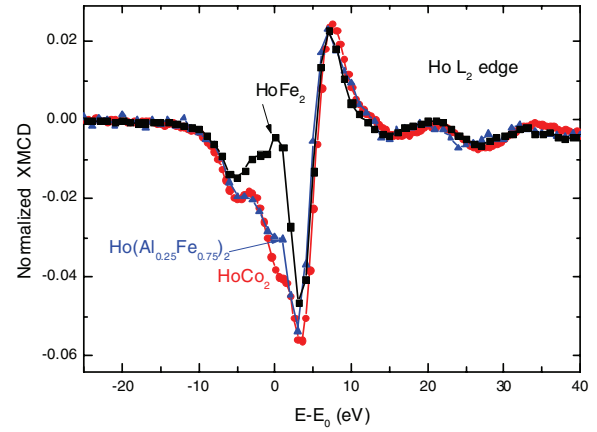
## RESULTS AND DISCUSSION

Fig. 1 shows the normalized XMCD spectra recorded at the rare-earth  $L_2$ -edge in both  $\text{R}(\text{Al}_{1-x}\text{Fe}_x)_2$  and  $\text{R}(\text{Al}_{1-x}\text{Co}_x)_2$  samples for  $R = \text{Ho}$ . Similar spectral behavior is observed when Fe Laves phases with different heavy rare-earth are measured [5]. The spectral profile of  $\text{HoAl}_2$  consists of a main negative peak centered at  $\sim 2$  eV above the edge and a positive peak at higher energy,  $\sim 7$  eV. In addition, Ho spectra present a negative shoulder at  $\sim -5$  eV. When T is placed in the lattice, the spectrum height undergoes a decrease. This reduction does not occur equally throughout the whole energy range. As the T content increases, the low energy part of the main negative peak undergoes an abrupt decrease, while the diminution in the higher energy part is much slighter.



**FIGURE 1.** Comparison of the Ho  $L_2$ -edge normalized XMCD signals measured at  $T = 5$  K and  $H = 5$  T for  $\text{Ho}(\text{Al}_{1-x}\text{Co}_x)_2$  (right panel) and  $\text{Ho}(\text{Al}_{1-x}\text{Fe}_x)_2$  (left panel) samples.

The profile change when the Al is replaced by a magnetic 3d metal is similar for both series. In both cases, reduction is more noticeable in the region of the XMCD spectrum around 0 eV. This large decrease at  $\sim 0$  eV can be regarded as the result of a positive peak emerging as the T content increases, which superimposes to the  $\text{HoAl}_2$  signal. The only difference between the XMCD spectral profile of both series concerns the intensity of the newly emerging peak. Thus, the intensity of the XMCD spectrum of  $\text{RCO}_2$  at  $\sim 0$  eV is clearly smaller than that of  $\text{RFe}_2$  compounds, while is more similar to that found in compounds with 75% of Fe content. This comparison is clearly seen in Fig. 2.



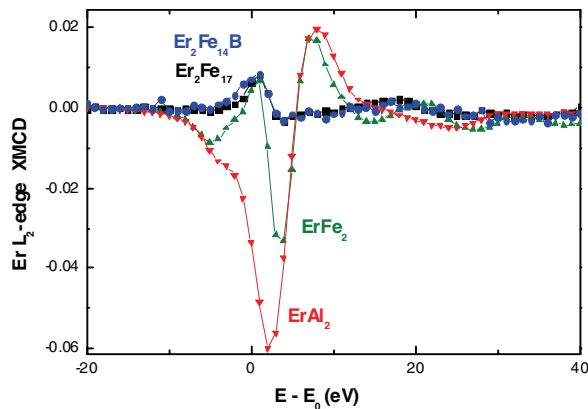
**FIGURE 2.** The effect of T substitution is much stronger in the case of Fe, as shows the comparison of the Ho  $L_2$ -edge normalized XMCD signals measured in  $\text{Ho}(\text{Al}_{0.27}\text{Fe}_{0.75})_2$ ,  $\text{HoFe}_2$  and  $\text{HoCo}_2$  samples.

Since substitution of Al by T does not modify the crystal structure, this evolution of the spectra shape has been already associated to the presence of Fe atoms, and more particularly to the number of Fe neighboring atoms. The present results on Co compounds verify, on the one hand, that the magnetic 3d metal is at the origin of an extra contribution to the rare-earth  $L_2$ -edge XMCD spectra. The explanation to this new emerging feature should be searched in the  $\text{R}(5d)\text{-T}(3d)$  hybridisation. Indeed, this can be regarded as the counterpart to the well known R contribution to the Fe K-edge XMCD spectra on R-T intermetallics [8]. In addition, the weaker Co effect indicates that the new feature is not only connected to the number of T neighbours, but also to the magnetic moment of the transition metal in the crystal lattice.

Finally, as the  $\text{R}(5d)\text{-T}(3d)$  hybridisation is not specific of the Laves phases, but it is considered to play an essential role in the magnetism of all the R-T intermetallic compounds, the influence of Fe can be expected to be present independently on the R:T

series. Thus, in Fig. 3 we present a comparison of the Er  $L_2$  XMCD spectra for 1:2, 2:14 and 2:17 series.  $\text{Er}_2\text{Fe}_{17}$  and  $\text{Er}_2\text{Fe}_{14}\text{B}$  spectra correspond to earlier measurements recorded at room temperature and under different applied magnetic fields [9]. Therefore, the intensity of the spectral features cannot be directly compared. However, the relative intensities of the main features for a given spectrum provide a clear confirmation of the presence of a contribution of Fe origin. It should be noted that in order to perform a correct comparison  $\text{Er}_2\text{Fe}_{17}$  and  $\text{Er}_2\text{Fe}_{14}\text{B}$  spectra have been multiplied by  $-1$ . This is due to the fact that the magnetization is dominated by the R-sublattice,  $M_R$ , in the Laves phases, while the contrary holds for  $\text{Er}_2\text{Fe}_{17}$  and  $\text{Er}_2\text{Fe}_{14}\text{B}$  being the Fe-magnetization,  $M_{\text{Fe}}$ , larger than  $M_R$ . The effect of the Fe sublattice is reflected not only in the sign of the XMCD spectra but also in the relative magnitude of the features. Thus, when Er is the only magnetic element, as in  $\text{ErAl}_2$ , the spectrum has a main negative peak at  $\sim 2$  eV whereas in  $\text{Er}_2\text{Fe}_{17}$  and  $\text{Er}_2\text{Fe}_{14}\text{B}$  this peak at  $\sim 2$  eV has almost disappeared. For these two compounds the main feature is a positive peak at  $\sim 0$  eV.  $\text{ErFe}_2$  presents an intermediate behaviour with both a positive peak at  $\sim 0$  eV and a negative peak at  $\sim 4$  eV.

According to our hypothesis, the magnitude of the feature at  $\sim 0$  eV is related to  $M_{\text{Fe}}$ . This explains why the spectra of  $\text{Er}_2\text{Fe}_{17}$  and  $\text{Er}_2\text{Fe}_{14}\text{B}$  present a profile with a main peak at  $\sim 0$  eV. Due to the large  $M_{\text{Fe}}$ , the Fe contribution at  $\sim 0$  eV is the largest feature and almost hides the R contribution. In addition, a very similar profile, is in agreement with a similar  $M_{\text{Fe}}:M_R$  ratio. On the other hand,  $\text{ErFe}_2$  has a relatively smaller Fe contribution to the XMCD signal so that both the negative and the positive peaks are similarly noticeable. Therefore, our results indicate that the existence of a magnetic T contribution to the R- $L_2$  XMCD spectrum is a general result for R-T alloys.



**FIGURE 3.** The effect of Fe: comparison of the Er  $L_2$ -edge XMCD signals measured for different series: 1:2, 2:14, 2:17. The signal corresponding to  $\text{ErAl}_2$ , without Fe content, has also been included for the sake of completeness.

## CONCLUSIONS

We have presented a systematic XMCD study at the rare-earth  $L_2$ -edge in  $\text{R}(\text{Al}_{1-x}\text{T}_x)_2$  compounds. Our aim was to disentangle which information can be gained from the XMCD spectra at this edge by modifying both the magnetic T atom and the R:T content ratio. The modifications observed when changing any of the above parameters point out the presence of a contribution associated to the magnetic 3d metal. The origin of this extra contribution can be addressed to the R (5d)-T(3d) hybridization. Furthermore, our results point out that the magnitude of the new emerging feature resembles the magnetization of the T sublattice. In addition, the measurements performed on the 2:17 and 2:14 series indicate that this contribution is a general characteristic in R-T compounds. Therefore, our present results point out the need of including an iron (cobalt) contribution prior to account for the XMCD at the rare-earth  $L_{2,3}$ -edges in R-Fe (Co) intermetallic materials in agreement with theoretical work [4].

## ACKNOWLEDGMENTS

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